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To: Dr. D. Mills
Director of Research

APPLICATIONS OF LASERS TO CANDU NUCLEAR FUEL REPROCESSING

K. B. Woodall, L. Mannik and H. D. Morrison

As part of a program to assist AECL in the development of CANDU Nuclear fuel reprocessing, we reviewed the application of lasers to reduce the costs or to improve the safety and efficiency of reprocessing spent nuclear fuel. Significant cost reductions may be possible using lasers to declad nuclear fuel. Other applications for lasers include monitoring of iodine-129 in gaseous effluents and monitoring of plutonium valence states in the main process stream. A paper summarizing our work up to the time AECL decided to withdraw from reprocessing research is attached. This paper was presented at the American Nuclear Society Topical Meeting on Waste Management and Decontamination and Decommissioning which took place in September, 1986, in Niagara Falls, New York. The paper will be published by the American Nuclear Society in 1987.

INTRODUCTION

The American Nuclear Society Topical Meeting on Waste Management and Decontamination and Decommissioning was attended by about 650 engineers and scientists from 17 countries including Canada, the U.S., Japan, China, and most of the countries of Western Europe. The meeting was strongly supported by U.S. National Labs and the Department of Energy, U.S. consultants and suppliers, and by all the countries of Europe and Asia with strong nuclear programs. AECL and Ontario Hydro were well represented and made more than a half dozen presentations but the conference was poorly attended by U.S. Utilities who operate nuclear reactors.

Our review of laser applications in Candu fuel reprocessing identified a large number of potential laser applications. Fuel decladding showed the most promise for cost reduction. Lasers may also make an important contribution in on-line high sensitivity monitoring of

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process parameters. Recovery of valuable isotopes or elements other than plutonium from spent fuel is a major potential application for lasers but this application was not pursued in any detail because by-product recovery is not an essential part of the plutonium recovery process.

Ontario Hydro was not the only research group attending the conference to consider lasers for use in remotely processing nuclear wastes. B. Meredith of the UKAEA gave a presentation on the use of carbon dioxide lasers in the size reduction of solid alpha wastes and slides of carbon dioxide lasers used in remote cutting operations at the Oak Ridge fuel reprocessing plant were shown as part of a talk describing all the remote materials handling facilities at Oak Ridge.

A.F. Baljet
Manager
Electrical Research Department

K.B. Woodall
Engineering Physicist
Science Section
Electrical Research Department

L. Mannik
Engineering Physicist
Science Section
Electrical Research Department

H.D. Morrison
Engineering Physicist
Science Section
Electrical Research Department

APPENDIX

APPLICATION OF LASERS TO CANDU NUCLEAR FUEL REPROCESSING

K. B. Woodall, L. Mannik, and H.D. Morrison

Ontario Hydro Research Division
Toronto, Ontario

ABSTRACT

A study has been made of the application of lasers to CANDU nuclear fuel reprocessing. We looked for ways to reduce the costs or to improve the safety and efficiency of reprocessing spent nuclear fuel. Among the areas adopted for further investigation were fuel decladding and process monitoring. These studies were initiated as part of a program of assistance to AECL in which the goal was to develop a reprocessing procedure specifically for CANDU fuel which would take advantage of the lower burnup and the lower levels of fissionable materials in the fuel in order to reduce costs.

INTRODUCTION

We have reviewed Atomic Energy of Canada Limited (AECL)'s proposed flowsheets for CANDU fuel reprocessing and have identified several areas where lasers could be used to reduce the costs or improve the safety and efficiency of reprocessing. Areas investigated include laser decladding and the monitoring of low level components in process and waste streams. We also looked at the use of light to replace the need for chemicals in uranium oxidation and in plutonium reduction and at laser driven by-product recovery from spent nuclear fuel. As decladding and monitoring held the most promise of improving fuel reprocessing, they were studied in more detail. Although light can be used to alter the oxidation states of uranium and plutonium, the technical difficulties of delivering light to the opaque liquids of that part of the reprocessing flowsheet made this application less attractive than several others. Even though the recovery of valuable by-products from spent fuel could, in principle, offset some reprocessing costs and simplify nuclear waste disposal, we did not study this option in any detail because by-product recovery is not an essential feature of any reprocessing flowsheet.

FUEL DECLADDING

Background

In light water reactor fuel reprocessing, a large hydraulic shear is used to chop up fuel rods to expose the fuel so that it can be dissolved prior to plutonium and uranium recovery. Blade replacement is frequent

requiring an elaborate, complex, and expensive remote maintenance system. Shearing of fuel rods leads to "pillowing" (partial closing of fuel rod ends) and substantial dust generation. Grid spacers and other subassembly parts can also jam the shear(1,2).

In laser decladding, a laser beam replaces the blade of the shear. Because this is a non-contacting method of cutting, no forces are exerted on the fuel and there is no tool wear. The main advantage of laser decladding is that much of the high technology equipment requiring maintenance can be located outside the hot cell.

Lasers are being used either experimentally or routinely to disassemble fast reactor fuel in Britain(3-7), France(8), and the United States(9-11). They are being tested experimentally by the United Kingdom Nuclear Power Development Authority for the decladding of LWR fuel at Springfields Nuclear Power Development Laboratories(6) and are being used routinely by Westinghouse(10) to recover nuclear fuel pellets from fuel bundles that fail quality control tests. Also in the US, Rockwell and Oak Ridge are assessing the use of lasers for the decladding of fast reactor fuel.

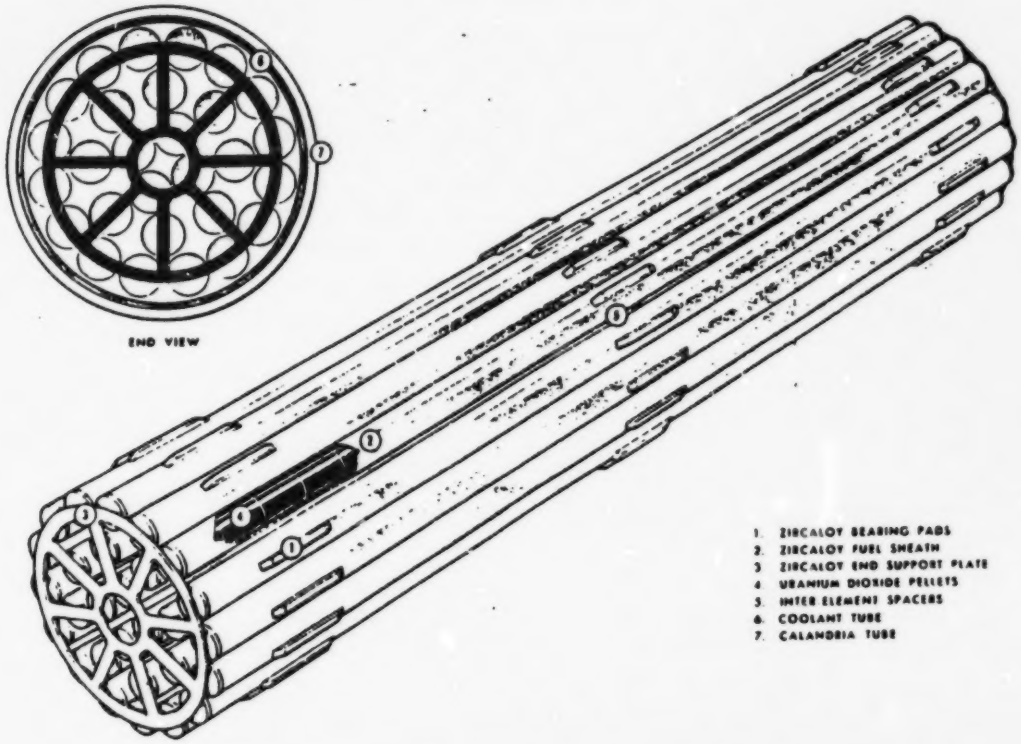

Figure 1. A CANDU reactor fuel bundle.

CANDU fuel (Figure 1) lends itself particularly well to laser decladding. The fuel bundles are small (about 50 cm long by 15 cm in diameter). Instead of containing a large shear and decontamination equipment, the hot cell need be little bigger than the size of the fuel bundle and the automated workpiece motion system. Light is transmitted into the hot cell

by either an optical fiber (YAG laser) or a ZnSe window (CO_2 laser). YAG lasers cost more per watt than CO_2 lasers but their energy is more effectively absorbed by zircaloy cladding. In preliminary cutting tests performed for us at Raytheon(12), 90 W of YAG laser power cut zircaloy cladding at the rate of 2 cm/second. Even faster cutting rates are expected at this power level but these preliminary tests did not attempt to measure maximum cutting rates. About twice as much power would be required if a carbon dioxide laser were used for this purpose but this would not increase the capital cost of the laser cutting system.

CANDU fuel bundles typically contain from 28 to 35 fuel elements arranged in concentric circles. Each element is about 12 mm in diameter. During fabrication the uranium dioxide fuel pellets slide easily into the fuel elements. They are not bonded to the zircaloy fuel sheath. Each fuel element is welded to two end support plates. The diameter of each of these welds is about 2 mm.

Laser Decladding

In order to dissolve the spent fuel prior to reprocessing, the zircaloy cladding which is about 0.38 mm (0.015 inches) thick must be cut. Because of the geometrical arrangement shown in Figure 1, a laser cannot easily access the inner fuel elements without the prior removal of the outer elements. Any laser decladding process must therefore involve two steps. First the fuel bundle must be separated into individual fuel elements and then the laser must cut into the zircaloy cladding in each fuel element.

Separating the fuel elements can be accomplished in two ways. Lasers can either cut the welds or cut the end plates. Cutting the welds involves directing two laser beams toward the side of the bundle. The focus of each laser beam is adjusted by a cam that simultaneously contacts both the fuel rod and the end plate in order to locate the exact position of each weld. For a 28 element fuel bundle with each weld taken to be a piece of zircaloy 2 mm x 2 mm x 2 mm, 11.2 cm of 2 mm thick zircaloy must be cut. Alternately to cut the 1.5 mm thick end plates we must make 72 cuts one cm long for a total cutting length of 72 cm in 1.5 mm thick zircaloy. In this case the laser focus is controlled by a cam contacting the end plate. Non-contacting sensors could also be used to determine workpiece position in order to set the laser focus. This can be done using optical techniques or even by measuring the pressure in an open ended tube containing flowing gas and facing the workpiece.

Laser cutting rates for zircaloy are extremely fast. For example, a 3 mm thick sheet can be cut at a rate of 6 m/min or 10 cm/sec by a 500 W carbon dioxide laser(13). This is more than double the cutting rate for steel. The same laser would cut 2 mm zircaloy at about 10 m/min (16 cm/sec) and 1.5 mm zircaloy at about 12 m/min or 20 cm/sec. A 200 W YAG laser beam will cut 1.5 mm thick zircaloy at the rate of about 7.7 m/min or 13 cm/sec. Cutting the welds with two 200 W YAG beams or two 500 W carbon dioxide laser beams would take a total laser cutting time per fuel bundle of 0.35 or 0.25 seconds respectively. Allowing one second for the laser to focus on each fuel element we predict a weld cutting time of about 30 seconds per fuel bundle. Cutting the end plates with the same two YAG or carbon dioxide lasers would take 2.8 or 1.8 seconds respectively. In this approach, laser

repositioning time for all the cuts would be unlikely to exceed 10 seconds. In each of the above approaches it may be necessary to catch each separated fuel element with a robot arm and place it on a support tray for quick retrieval. This is estimated to require a processing time of 30 seconds per fuel bundle regardless of which type of laser cutting is used.

Having separated the fuel elements the next step is to expose the fuel by cutting into the zircaloy cladding. To do this the output from two 400 W YAG lasers or from one 1500 W carbon dioxide laser is split into eight equal beams in an optical system external to the hot cell. A robot arm would grasp each element in the center and spin it through one rotation while eight equally spaced circular cuts about 6 cm apart are made. Cutting times of about 0.15 seconds per fuel element are estimated. It should be possible to process each element in about a second for a processing time of about 30 seconds per fuel bundle. The total processing time for both separating the fuel elements and decladding is estimated to be one minute per fuel bundle. Note that the laser cutting time will be a small fraction of this minute.

The laser beams that do the cutting are tightly focused. The higher melting uranium oxide fuel beneath the cladding will not be melted or vaporized by the laser beam. When a laser beam cuts zircaloy, part of the material is vaporized and part is liquefied and removed as a liquid. In our trial tests cutting empty tubes the removed material solidified into small particles at the opposite side of the tube from the cut. When cutting fueled tubes, we may find it necessary to put a suction and filter device around each laser nozzle to capture the small amount of removed material.

Cost Estimates and Future Work

A 1.5 kW carbon dioxide laser costs about $120,000 and two 400 W YAG lasers would cost $160,000 to $200,000. The carbon dioxide laser appears at this time to be the lowest cost option. Since automation systems generally cost one to two times the cost of the laser(14), the workpiece motion systems are expected to cost between $120,000 and $240,000. Hydro's Design and Development Division has told us that a complete French mechanical shearing system and hot cell for LWR fuel costs about $25 million. With our first estimate of the cost of the laser system near 1% of this cost and with a hot cell only about three times the size of the very small CANDU fuel bundle, there is an excellent chance that decladding CANDU fuel could be done for less than 10% of the cost of a shearing system. We are planning to acquire a 1.5 kW carbon dioxide laser system that could be used to demonstrate CANDU fuel decladding. This laser will enable us to improve our cost estimates for the laser decladding process and also enable us to demonstrate shorter term cutting and welding applications within Ontario Hydro.

PROCESS MONITORING

The most important monitoring needs that have been identified are the monitoring of low levels of iodine-129 in effluent gases and the monitoring of plutonium valence states.

Iodine-129 Monitoring

Background

Neglecting U-238, iodine makes up 0.36% by weight of spent nuclear fuel from CANDU reactors(14,15). About 80% of the iodine is radioactive iodine-129, which has a half-life of 1.6×10^7 years, and the rest is stable iodine-127. (The expected 5-year storage period for spent fuel ensures that all the other iodine isotopes have decayed to negligible quantities.) Fuel reprocessing releases iodine as a gas. Prior to discharging the effluent gases to the atmosphere, most of the iodine will be removed by a silver impregnated solid sorbent filter. Measurements of iodine-129 concentration are required before and after filtering, with typical concentrations ranging from 30 ppm before the filter to 100 ppb after the filter.

Since conventional counting techniques are impractical due to the low activity of iodine-129, attention has turned to optical methods, which are well suited to on-line, real-time monitoring applications. Earlier work by Vikis at WNRE(16) explored several laser-based techniques for detecting iodine, including optical absorption and laser induced fluorescence (LIF) using a helium-neon (He-Ne) laser. The best sensitivity was obtained with an intracavity LIF method which yielded a detection limit of 1 ppb. However, none of these techniques is adequate in the presence of NO_2, which is present at the 2 to 3% level and which also fluoresces under He-Ne laser excitation.

Similar problems were encountered by researchers in the US. A group at Naval Research Laboratory developed a prototype iodine-129 detector using LIF excited by an isotopic helium-neon laser (^3He-^{22}Ne)(17). This gave a detection limit of 10 ppb but again the presence of NO_2, in this case at the 0.3% level, represented a serious interference with iodine-129 measurements at concentrations below 300 ppb.

The aim of the present research was to again explore LIF as a detection technique, since it represents a highly sensitive method that lends itself to remote, on-line monitoring, but to avoid NO_2 interference by selecting an excitation wavelength where I_2 fluoresces but NO_2 does not. Since there was reason to believe that I_2 would fluoresce over most of the visible and ultraviolet spectral range, the study focused on the ultraviolet region near 250 nm where NO_2 fluorescence is expected to be minimal. This is because NO_2 has a very low absorption coefficient at this wavelength and absorption is a prerequisite for fluorescence.

Ultraviolet Laser Induced Fluorescence of Iodine

The Quantel Nd:YAG pumped dye laser has a tuning range of 295 to 750 nm. Previous experience in our laboratory with extending the tuning range into the infrared using non-linear crystal mixing techniques(18) proved useful in the present conversion into the ultraviolet using similar methods.

Figure 2 is a schematic of the ultraviolet laser design. The Nd:YAG laser operates at a fixed wavelength of 1.06 microns which is frequency doubled to 532 nm. The remaining 1.06 micron beam is optically delayed using a set of mirrors and the appropriate polarization for mixing is

selected using quarter-wave plates. The green 532 nm beam pumps the dye laser (rhodamine 640 dye) to generate tunable output in the red near 654 nm which is frequency doubled to yield ultraviolet output near 327 nm. A glass filter rejects the 654 nm beam and passes the 327 nm and 1.06 micron beams which are combined in a crystal of KDP (potassium dihydrogen phosphate). By angle tuning this crystal, it is possible to generate output near 250 nm by the process of sum frequency mixing; ie, 1.06 microns (9,434 cm^{-1}) + 327 nm (30,566 cm^{-1}) produces uv radiation at 250 nm (40,000 cm^{-1}). The three beams emerging from the mixing crystal are spatially separated by a prism and the 250 nm beam, whose wavelength is adjustable by tuning the dye laser, is passed into the iodine fluorescence cell. In this way, pulse energies of 0.5 to 1 mJ were generated in the ultraviolet.

An all-quartz cell consisting of three quartz windows fused onto the cell body was constructed for these measurements(19). This design is preferable to the simpler approach of sealing windows onto the cell because iodine vapor attacks most sealing compounds(20).

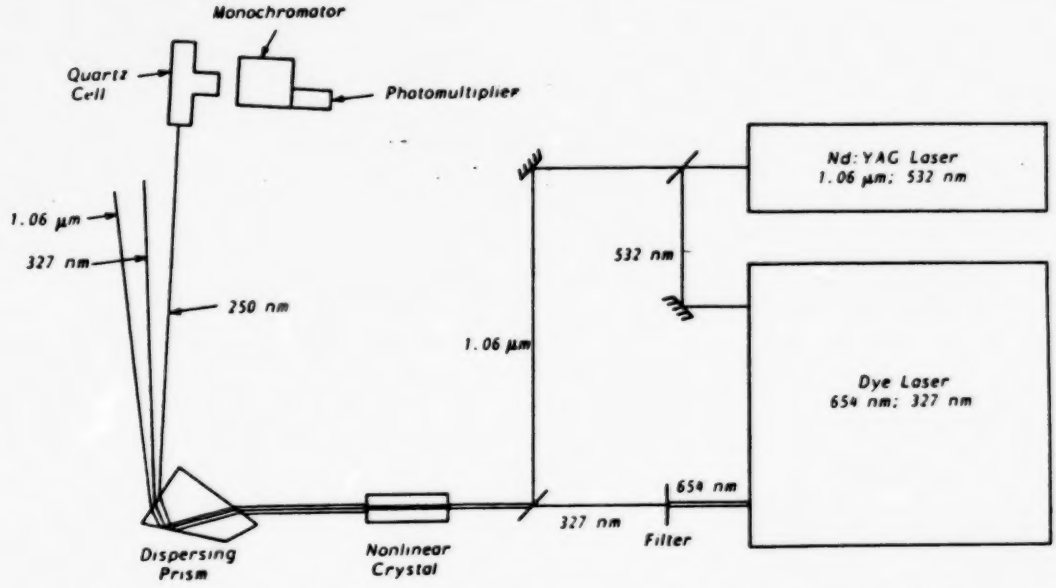

Figure 2. Generation of Tunable Ultraviolet Output For Iodine-129 Monitoring Using Nonlinear Mixing.

Iodine crystals were placed in a finger attached to the bottom of the fluorescence cell. At room temperature (23°C) the vapor pressure of iodine, as calculated from an equation in reference 20, is 284 mtorr.

The iodine fluorescence passed through a window at 90° to the laser beam and was spectrally analyzed with a monochromator (Monospec) connected to a photomultiplier detector (Hamamatsu R446). The monochromator was scanned while recording the photomultiplier output on a strip chart recorder, using a lock-in amplifier to improve the signal to noise.

With ultraviolet excitation the only signals detected were a sharp line at the excitation wavelength and a broad, featureless emission at higher wavelengths. These were ascribed to scattering and fluorescence originating in the quartz input window and transferred along the wall of the cell to the viewing window. It is certain that the observed fluorescence is not due to iodine since there was no change in the measured spectrum when the iodine was removed from the path of the laser beam by freezing out with liquid nitrogen.

There is one reference in the literature to earlier attempts to measure the fluorescence of iodine using excitation near 250 nm provided by a low pressure mercury lamp(21). In that investigation, fluorescence was only detected at high temperature (500°C) and at a pressure of 100 torr and became intense only at atmospheric pressure (760 torr). Other studies of iodine fluorescence stimulated by vacuum ultraviolet excitation (below 200 nm) have demonstrated that the addition of a buffer gas increases the fluorescence intensity(22).

These observations suggest that one promising approach for enhancing the fluorescence of iodine near 250 nm would be to add enough nitrogen buffer to bring the total pressure up to one atmosphere and to operate at higher temperatures by enclosing the cell in an oven. Another advantage of using the buffer gas is that it would make the experimental conditions more representative of the situation in a reprocessing plant where trace amounts of iodine would be present in an atmosphere of air.

Alternative Approaches

As mentioned above, the fluorescence of iodine has been observed under vacuum ultraviolet excitation, specifically using an argon fluoride (ArF) laser at 193 nm(22). Since NO_2 is not expected to fluoresce in this spectral region, this represents one alternative approach that may be worth investigating. Excimer lasers such as ArF have undergone rapid development over the past several years and commercial units are currently available for $20,000.

Another iodine detection system that we have considered is two-photon spectroscopy(23). Research on this subject has been performed by J.R. Robins of our research group(20) using two tunable dye lasers to populate an upper level in two steps and measuring the subsequent emission from this upper level. This method can be readily adapted to detect any of the isotopic forms of iodine and would probably have adequate sensitivity. While at first glance the cost of two lasers may seem prohibitive, there are compact nitrogen laser pumped dye lasers on the market for as little as $5,000. It may also be possible to design the process so that both photons are generated by a single dye laser system.

Plutonium Valence States

Efficient processing of spent nuclear fuel requires measurement of the relative concentrations of plutonium valence states, Pu(III), Pu(IV), and Pu(VI), at various stages in the process. These concentrations are currently measured off-line with a relative accuracy of 5% using a uv-visible spectrometer. It is desirable to detect the Pu(III) and Pu(VI) states down

to 0.1% or 4×10^{-6} moles/l, the projected concentration in the head end of a CANDU fuel reprocessing plant. An on-line detection method for these plutonium valence states would improve process efficiency.

A recent report by Stumpe et al.(24) describes a laser technique that has demonstrated measurement of state-specific Pu concentrations to less than 10^{-7} moles per liter (equivalent to 0.0025%) We believe that this method can be converted to an on-line method which meets the required detection limit of 4×10^{-6} moles per litre. The technique used is known as photoacoustic spectroscopy. It employs a single beam from a pulsed tunable dye laser which passes through a sample cell containing solute and solvent, and a reference cell that only contains solvent. The absorption of light in the cells generates pressure (sound) waves that are detected by piezoelectric transducers. Greater or lesser absorption produces pressure waves of greater or lesser intensity. By scanning the wavelengths of the dye laser, a photoacoustic spectrum similar to the light absorption spectrum can be generated. The magnitude of the photoacoustic signal is directly proportional to the ion concentration and to the light absorption coefficient so that absolute concentrations can be obtained by a simple calibration procedure. The reference cell is employed to provide the background spectrum of the solvent. Subtraction of the reference spectrum from the sample spectrum eliminates the signal due to the solvent and increases the detection sensitivity of the dissolved ions. Sensitivities of 7×10^{-8} moles/litre have been reported for Pu(IV) and 3×10^{-8} moles/litre have been reported for Pu(VI) in aqueous solutions. Sensitivity data for Pu(III) have not yet been reported. This work is required before attempts to build an on-line device begin.

One important aspect of photoacoustic spectroscopy is that for ease of maintenance and for protection of the equipment from radiation both the laser and the signal processing electronics can be positioned remotely from the solvent cells. This feature could prove important in the development of an on-line measurement system. However, the problems of particulates suspended in process solvents and of environmental acoustical noise will need to be examined before this technique can be applied on-line. Particulates which scatter and absorb laser light can shorten the light penetration depth to a few millimeters and acoustical noise can interfere with the piezoelectric sensors. Even if these problems are found to preclude on-line operation, photoacoustic spectroscopy can provide much more sensitivity than conventional techniques.

BY-PRODUCT SEPARATION

By the year 2000 Ontario Hydro's spent nuclear fuel stockpile will contain elements and isotopes worth billions of dollars at current market prices. If any of these can be recovered economically from the waste streams, the sale of that material would reduce reprocessing costs. Many lanthanides and most actinides can, in principle, be separated merely by shining uv light on them(25,26). The light induces the actinide or lanthanide to change oxidation state. This can cause the ion to precipitate from solution or cause it to change from the organic phase to the inorganic phase (or vice versa) during solvent extraction. We have examined a modified Purex flowsheet (one of the approaches being evaluated for CANDU

fuel reprocessing) for streams that can take advantage of optical separation techniques. A potential problem with optical separation schemes is the as yet unknown impact of the suspended solids in these streams. As the ionic solutions pass from one stage of the process to the next, they become progressively clearer. Optical separation techniques may have to be restricted to side streams and later portions of the process stream. The absorption of uv radiation by tributyl phosphate is another potential problem. The actinides (except for uranium and plutonium) and the lanthanides which would be susceptible to optical separation methods are located mainly in the aqueous waste stream of the Purex process.

OTHER APPLICATIONS

We also examined the possibility of using optical techniques instead of chemicals to alter the oxidation states of uranium and plutonium right in the heart of the Purex flowsheet(26). The reservations concerning suspended solids and the the light absorption properties of tributyl phosphate mentioned above apply particularly strongly to this application.

We are currently demonstrating in our laboratory a laser method of removing tritium from waste water streams(27). The tritium in the water is exchanged with CF_3H and removed from the CF_3H stream by tuning a carbon dioxide laser to selectively dissociate the CF_3T containing molecules. The TF created by this dissociation process is readily separated from the undissociated CF_3H by absorption on a sodium fluoride bed. This work may also be applicable to cleaning up the waste water streams at reprocessing facilities.

CONCLUSIONS

We have examined several potential applications for lasers in CANDU fuel reprocessing and have identified three applications which could either reduce costs or improve safety and efficiency. Laser decladding is technically feasible and has the potential of significantly reducing reprocessing costs. Laser monitoring systems could also have the potential to increase sensitivity and provide on-line monitoring for iodine-129 and plutonium valence states but the development and implimentation of these techniques for a reprocessing plant will present many challenges.

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